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RADIOECOLOGICAL STUDIES

ON THE

COLUMBIA RIVER

Part I

by

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C. E. Cushing
C. C. Coutant
W. L. Templeton

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Part II of this report, Appendices A and B, contain the tabular data of all analyses and is available from the authors on request.

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RADIOECOLOGICAL STUDIES ON THE COLUMBIA RIVER

PART I

May 1970

AEC RESEARCH & DEVELOPMENT REPORT

BNWL-1377

BATTELLE NORTHWEST

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Figures and Tables

ERRATA

1. The list of Figure and Table titles, following the Table of Contents, is bound in reverse order.

2. In the Figure title for Figure 12 on the above list, zirconium-niobium-94 should read zirconium-niobium-95.

A historical account of earlier radioecological studies together with a basic description of the Columbia River, its chemical and physical characteristics, and the extent of the drainage basin have been reported (3, 4).

Changes modifying river conditions have taken place since the studies conducted before 1956 and those of this report which were conducted between February 1966 and September 1967. The number and location of the operating reactors have changed with time (Fig. 1). As many as eight reactors were in operation during the studies prior to 1956. In late 1964 and early 1965 three reactors closed, and a fourth was shut down during the course of this investigation in 1967. The effect of the early reactor closures on the concentrations of radionuclides in the river biota has been reported (9). Six reactors were operating during most of this study. One of these, 100-N, has a closed primary cooling system with river water used as a secondary coolant. Effluent from this reactor normally contains no radioactive materials, but does introduce heat into the river.

During this study the discharge of radionuclides into the river was interrupted for more than 40 days in July and August of 1966 when all reactors were shut down during a labor dispute. This was the first time since the start of plant operation in 1944 that all reactors were simultaneously out of operation. This disrupted the study of seasonal cycles of radionuclide concentrations in the river organisms, but provided a unique opportunity to observe the decline and subsequent accumulation of radioactivity in the biota during reactor closure and resumption of operation (11, 12). This reactor closure necessitated an extension of this study into 1967 to measure the "normal" seasonal values during the summer

RADIOECOLOGICAL STUDIES

IN THE

COLUMBIA RIVER

INTRODUCTION

The use of Columbia River water as a reactor coolant and the subsequent discharge of this cooling water into the River introduces a number of radioactive elements into the river environment. These radionuclides are produced by the neutron activation of stable elements in solution in the cooling water and the sloughing off of radioactive corrosion products from the surfaces of the reactor cooling tubes. The relationships of these radioactive materials to river organisms have been studied since reactor operations began in 1944 (1-10). Radioanalyses in the earlier studies (3, 4), dealing mainly with seasonal variations, species differences, and geographical distribution of radioisotopes by the river biota, were confined to the measurement of total beta emitter activity and to the estimation of relative amounts of phosphorus-32 through decay curve analysis.

The purpose of this study is (1) to define the interspecies and seasonal variations in the concentration of several of the more biologically important radionuclides, and (2) to update the findings of some of the earlier investigations.

period.

The establishment of hydroelectric dams upstream from Hanford has modified the river environment. Between 1956 and 1967 three low-head dams were built at distances from 20 to 147 km upstream from the reactors. Priest Rapids Dam, immediately upstream from the Hanford Reservation, exerts the greatest influence on the local river conditions. River plankton species composition and quantity have changed markedly (13). Dominant species are lentic rather than lotic. The retention and release of water at Priest Rapids, to satisfy varying daily electrical power demands, produces average diurnal river elevation fluctuations of 1.8 m, with maximum as high as 3.8 m immediately downstream (Fig. 2). Daily fluctuations of 2 m were common near our sampling sites.

Diurnal changes in river flow were correspondingly great, with the maximum often more than double the minimum. A low-head dam, such as that at Priest Rapids, exerts its greatest influence during average or low flow periods (fall and winter), and least during the freshet (spring and early summer).

These changes in hydrographic conditions have destroyed much of the littoral zone productivity. Near-shore production of benthic forms, particularly sessile algae, has been reduced due to restrictions in light penetration resulting from higher turbidity and greater mean water depth because of diurnal water level fluctuations. The stability of shoreline habitat used as feeding and resting areas by immature fish is destroyed. The rapid variations in river elevation have nearly eliminated organisms such as crayfish and water boatmen (Corixidae) from littoral zone areas of former abundance.

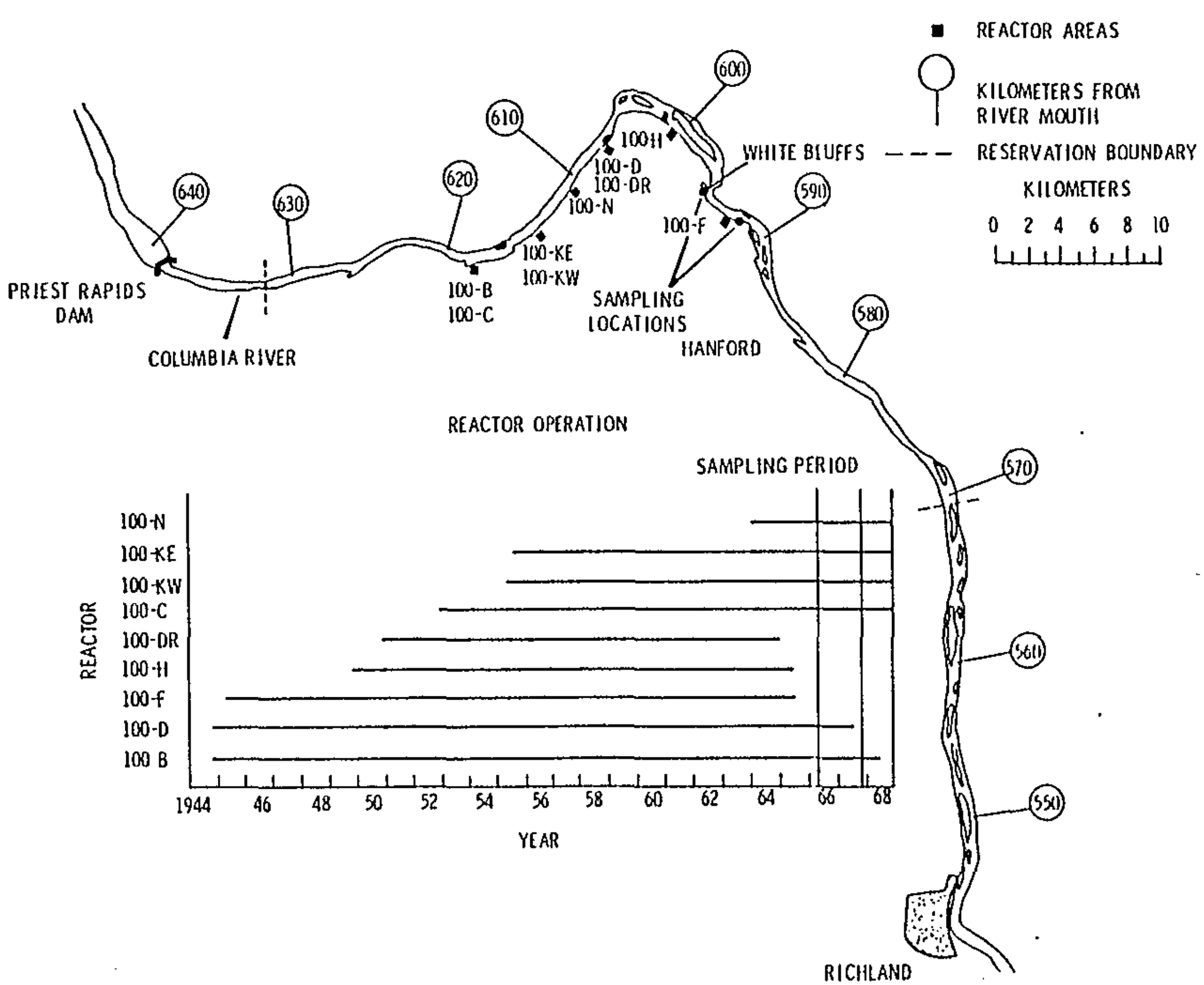

FIGURE 1. HANFORD REACTOR OPERATION AND LOCATION

METHODS

All samples, except large fish, were collected near 100-F area (Fig. 1), about 13.5 km downstream from the nearest reactor effluent outfall in a zone of nearly complete effluent mixing. The sampling site was characterized by a gently sloping bottom, substrate ranging in size from fine sediment to rubble 4 dm in diameter (average approximately 1 dm), and water depth fluctuating daily from 0.5 to more than 2 m.

Samples were obtained by wading during morning low water periods. Plankton was collected with 70 µ mesh nets suspended in the river currents, periphyton and invertebrates were picked or scraped from stones from the bottom, and small fish were collected with a hand seine in small inlets along shore. All samples were usually processed and submitted for radioactivity measurement within 2 hr after collection.

Large fish were collected near White Bluffs in an area about 11 km downstream from the nearest reactor where low current velocities permitted the use of gill nets. They were refrigerated until processed in the laboratory, usually on the day of collection.

Sample processing was as follows:

(1) Periphyton, sponge, caddisfly larvae - washed in river water, debris removed, excess moisture blotted with paper towels.

(2) Plankton - water decanted from settled sample, excess moisture blotted with paper towels.

(3) To standardize the removal of surface moisture, the above samples were placed in 20 ml plastic tubes with a water

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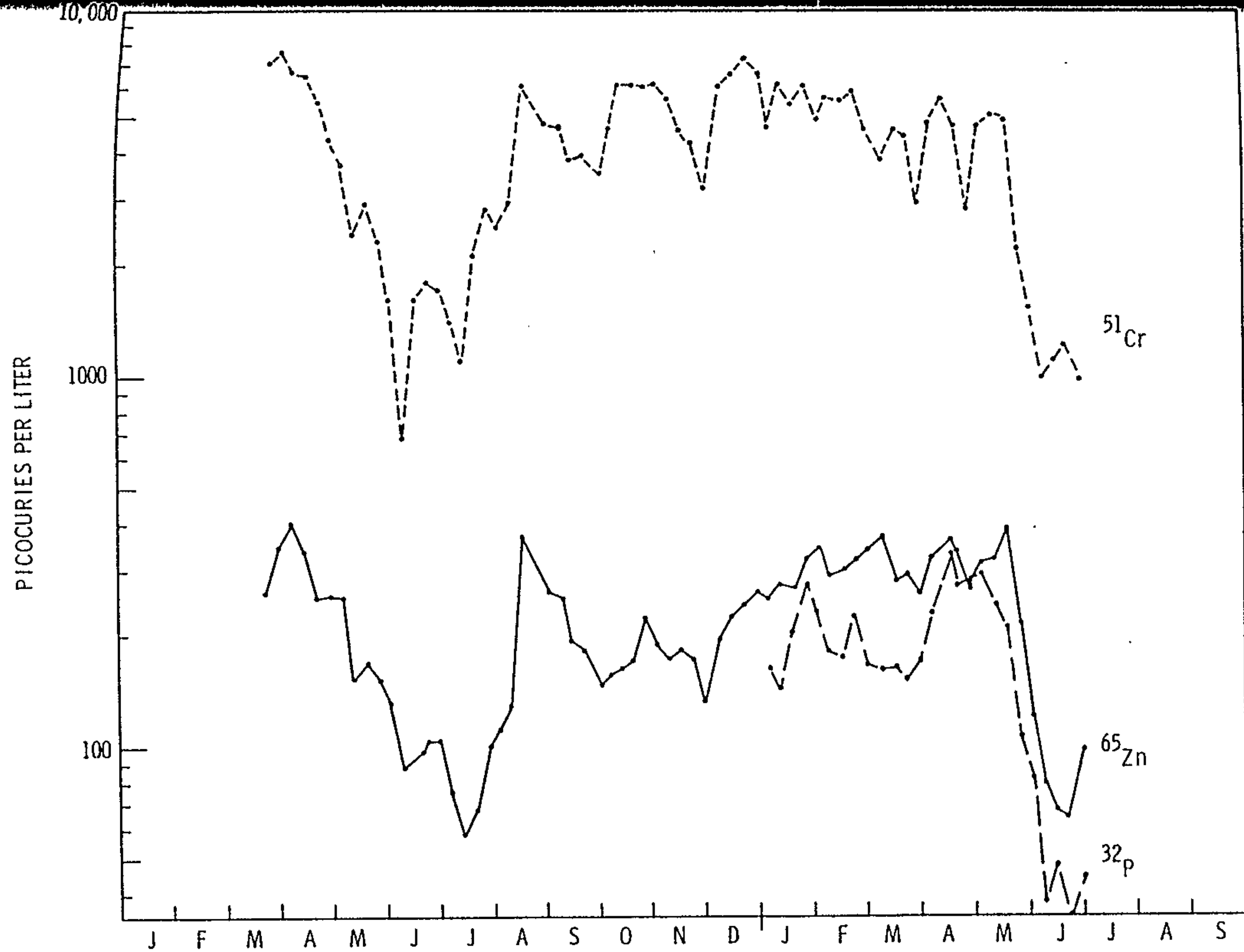

FIGURE 2. CONCENTRATION OF RADIONUCLIDES IN COLUMBIA RIVER WATER

and neptunium-239. Data transferred to punch cards for computer analyses.

(4) Initial computation of concentrations of gamma emitters was made by use of the GEM program (15); further analyses were made on output from a FORTRAN program.

Beta emitter analyses:

(1) Total beta - a single aliquot of 1 to 1.5 g wet wt spread evenly and dried on a 1" stainless steel planchett, counted with a wide beta, thin window, gas flow proportional analyzer.

(2) Phosphorus-32 - measured by a differential absorber technique (14), on the same sample and counting system used for total beta. A decay period of 7 to 10 days allowed before counting for phosphorus-32 for decay of interfering short-lived radionuclides.

(3) Results were calculated on a special computer program.

Standard weights:

(1) One to five grams wet sample taken on each series of samples, dried at 60 C and 15" of mercury vacuum for 24 hr for standard dry weight determination.

(2) Sample then ashed at 425 C for a minimum of 24 hr for determination of ash weight.

(3) All samples cooled to room temperature in a dessicator before weighing.

reservoir in the bottom and centrifuged for 5 min; caddis fly larvae at 700 rpm, remainder at 2000 rpm.

(4) Limpets - sediment and algae washed from shell, steamed over a water bath for about 3 min, shell and soft parts separated.

(5) Small fish - surface moisture blotted, gut contents removed, mascerated to form a homogenate.

(6) Large fish (at least 6 individuals of each species sampled when available) - gut contents removed, sample from anterior gut retained for radioanalysis, fish separated into muscle and "carcass" (remainder) fractions, ground to a homogenate.

Gamma emitter analyses:

(1) Plankton, periphyton, sponge, caddisfly larvae, limpets, small fish, and large fish gut contents - three aliquots of about 4 ml equivalent volume placed in the bottom of 20 ml glass test tubes, 1 ml 10% formalin added to inhibit decay.

(2) Large fish muscle and "carcass" - a 125 to 500 ml equivalent volume of each tissue from each fish put in 500 ml polyethylene bottle.

(3) Gamma measurement done with either a 3" x 3", or 9" x 11" NaI well crystal with readout on a 400 channel analyzer. Measurements were made for the following radionuclides: sodium-24, scandium-46, chromium-51, manganese-54, iron-59, cobalt-60, zinc-65, zirconium-niobium-95, ruthenium-rhodium-106, iodine-131, cesium-137, barium-140, lanthanum-140, cerium-praseodymium-144,

variations in water radioactivity are inversely related to river flow. This is apparent by comparison of seasonal trends in Fig. 2 with variations in river flow shown in Fig. 3.

Factors other than dilution may also influence the radionuclides in solution in river water. Zinc-65, manganese-54, zirconium-niobium-95, iron-59, and scandium-46 are associated mainly with particulates (17) and the concentrations of these isotopes in solution would be reduced by suspended silt in the River. These nuclides are depleted more rapidly with transport downstream than is chromium-51, for instance, which remains mainly in solution.

The concentrations of some radionuclides in water were slightly higher in 1967 than previously reported measurements. In September 1957, the zinc-65 and chromium-51 values were 89 pCi/l and 2000 pCi/l, respectively (7); in September 1967, they were 200 pCi/l and 4000 pCi/l, respectively. The 1957 measurements were made on a single grab sample and may be less representative than the analyses of integrated samples in 1967.

River organisms

Some of the 15 radionuclides for which analyses were made in the river biota were not present consistently and in sufficient amounts to be useful in demonstrating seasonal trends. Concentrations of phosphorus-32, zinc-65, chromium-51, manganese-54, iron-59, zirconium-niobium-95, and, in some instances, scandium-46 were measureable in most organisms throughout the year. To permit comparison with results of earlier

Stable element analyses:

Measured by flame spectroscopy (Na, K), colorimetric (P), and atomic absorption (Ca, Mg, Cu, Zn, Fe, Mn, Cr, and Co) techniques:

RESULTS AND DISCUSSION

Water

A number of radionuclides are introduced into the river in the reactor effluents but only a few are of biological significance. Some with short half-lives are quickly lost from the ecosystem through radioactive decay, while others are not accumulated to any extent by the river biota. Zinc-65 and phosphorus-32 are the radionuclides of greatest biological importance and chromium-51 is the most abundant. At Richland, about 45 km downstream from the reactors, chromium-51 accounts for approximately 50% of the total radioactivity in the water, and zinc-65 and phosphorus-32 contribute 2% and 1%, respectively (16).

The concentrations of chromium-51, zinc-65, and phosphorus-32 in water, collected with a continuous sampler near the organism sampling sites, are given in Fig. 2. The 1967 values for July through September replace the measurements for the same period in 1966 when the normal seasonal patterns were disrupted by the previously mentioned reactor closures. This replacement of one year's values for those of another is made with the realization that the precise pattern and quantity of radionuclides in the water may vary in consecutive years, but this approach is considered reasonable for the general illustration of seasonal trends. The effluent discharge rate is relatively constant so seasonal studies (3, 4), analyses were also made for total beta activity which will be discussed later. Sodium-24 was present in most biological materials, but its relatively short half-life (15 hrs), the lapse of several days between collection and lab processing and subsequent radioactivity counting, and the associated uncertainty of decay corrections, made the analytical results for this isotope questionable. Consequently they are not discussed in this report. The mean values of radionuclide concentrations in all samples are presented in Appendices A and B.

The seasonal variations in concentrations of phosphorus-32, zinc-65, chromium-51, manganese-54, iron-59, and zirconium-niobium-95 in representative river organisms are shown in Figs. 4 to 21. Similar to the treatment of the water data, the 1967 July through September biota values are substituted for the same values in 1966 when the reactor shutdown altered normal seasonal patterns. Values plotted on the graphs are the means of replicate analyses, when available. Values less than twice the associated counting and computation error were rejected and treated as zero in the calculation of the mean values. This tended to produce many zero values, shown as baseline points in the Figures, during seasons of the year when concentrations of certain radionuclides fluctuated around this cut-off point. This is particularly evident for the manganese-54 and zirconium-niobium-95 levels in net plankton (Figs. 5 and 6).

Variation in river flow, which produces changes in the dilution of the reactor effluents, and river temperature, which affects metabolic rate, are two of the most influential factors in the control of radionuclide content of the river biota. Comparison of river flow and

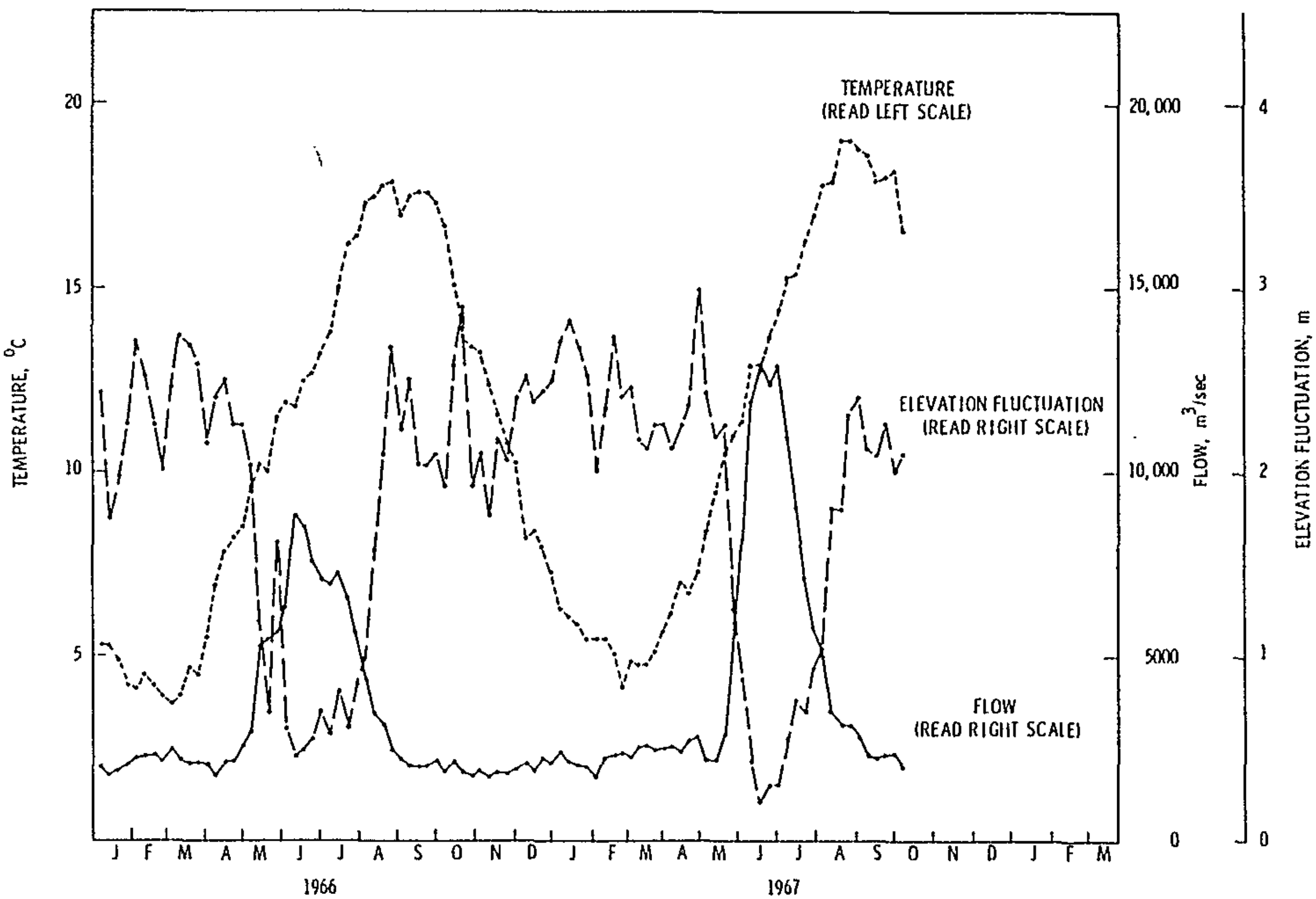

FIGURE 3. COLUMBIA RIVER MEAN WEEKLY TEMPERATURE, FLOW AND DIURNAL ELEVATION FLUCTUATION

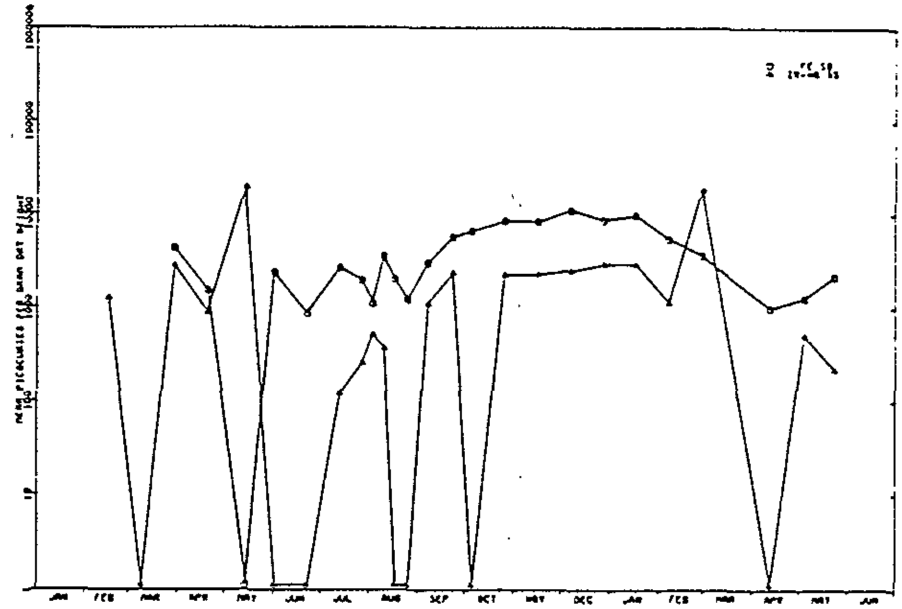

FIGURE 6. CONCENTRATIONS OF IRON-59 AND ZIRCONIUM-NIOBIUM-9
IN NET PLANKTON

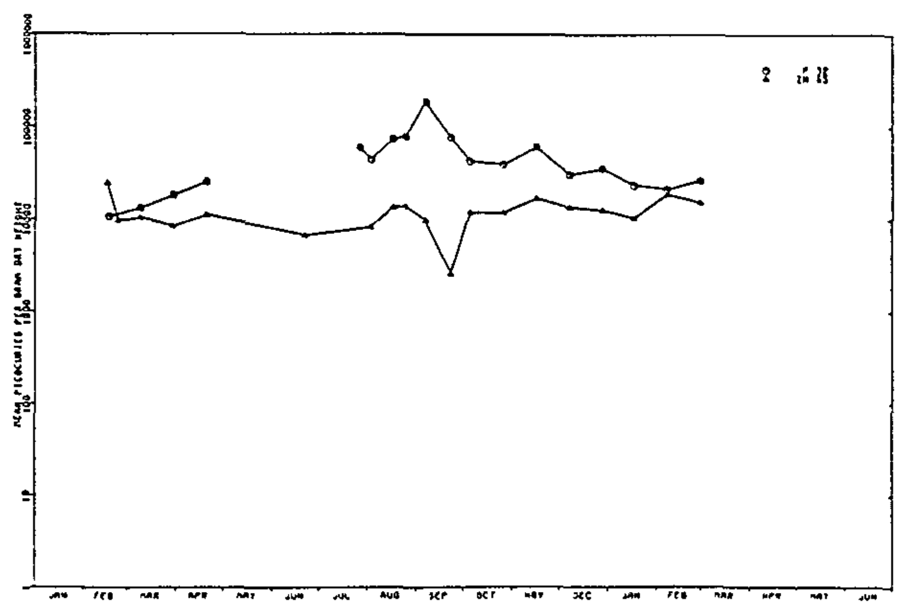

FIGURE 7. CONCENTRATIONS OF PHOSPHORUS-32 AND ZINC-65 IN PERIPHYTON

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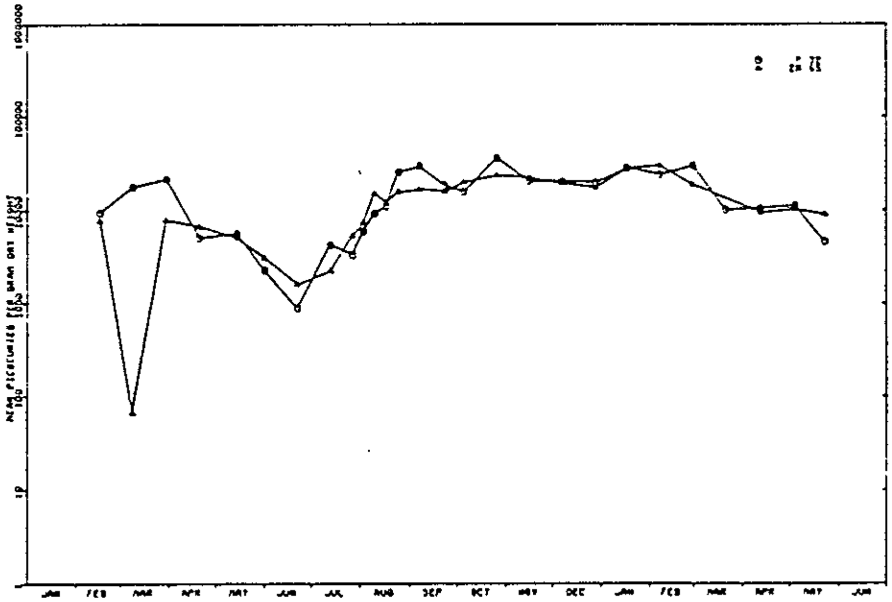

FIGURE 4. CONCENTRATIONS OF PHORPHORUS-32 AND ZINC-65 IN NET PLANKTON

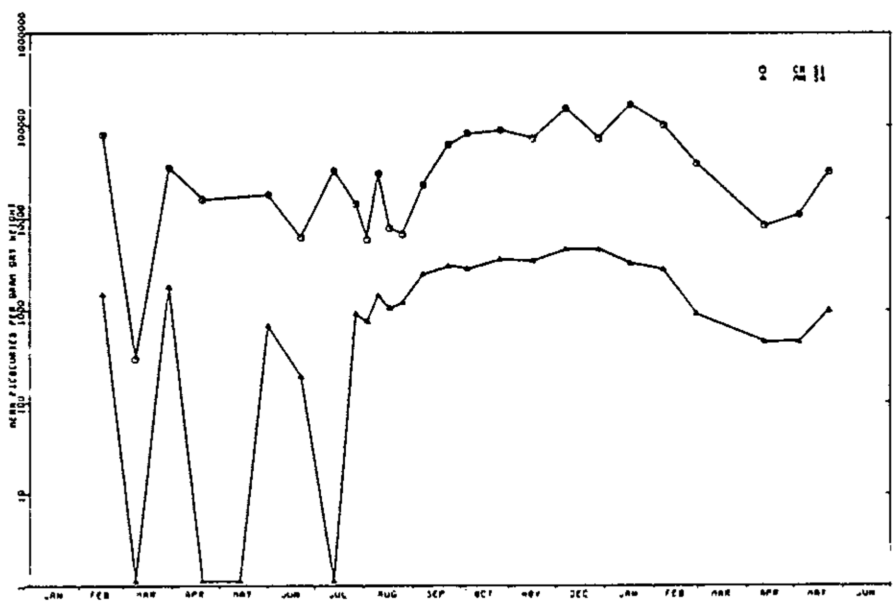

FIGURE 5. CONCENTRATIONS OF CHROMIUM-51 AND MANGANESE-54 IN NET PLANKTON

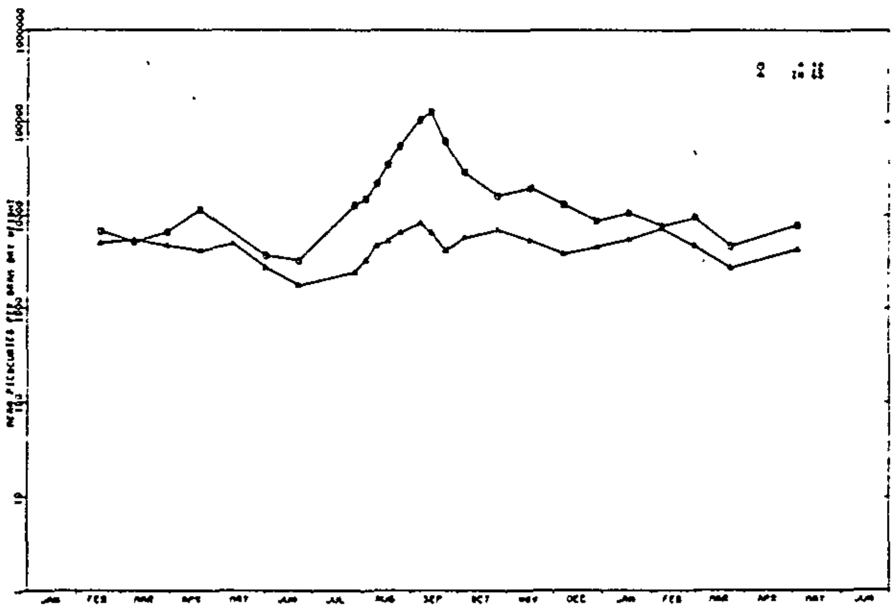

FIGURE 10. CONCENTRATIONS OF PHOSPHORUS-32 AND ZINC-65 IN CADDISFLY LARVAE, HYDROPSYCHIDAE

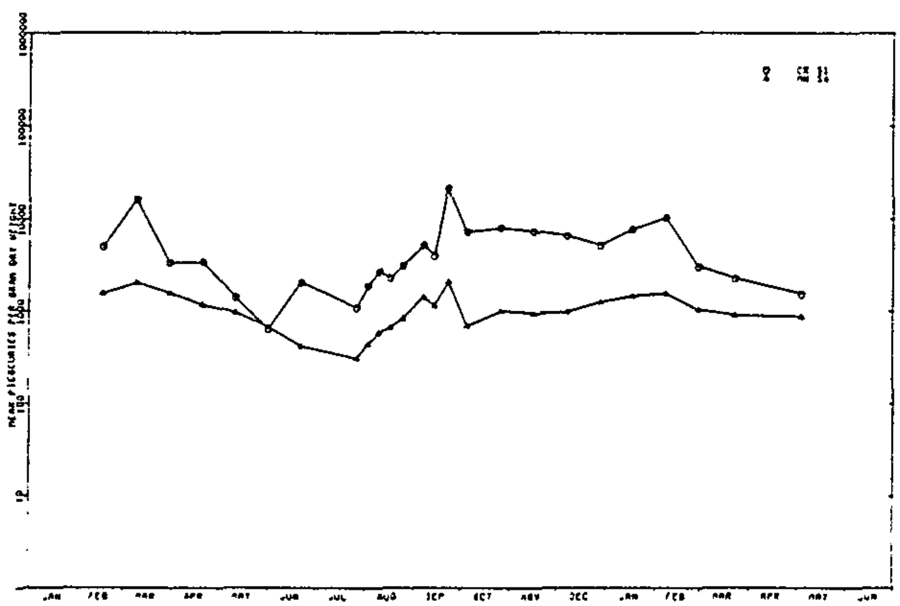

FIGURE 11. CONCENTRATIONS OF CHROMIUM-51 AND MANGANESE-54 IN CADDISFLY LARVAE, HYDROPSYCHIDAE

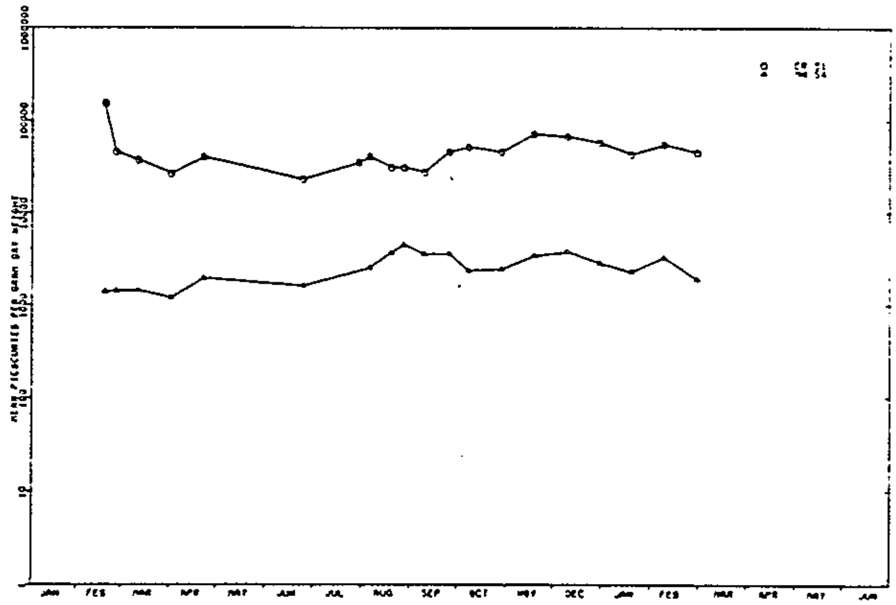

FIGURE 8. CONCENTRATIONS OF CHROMIUM-51 AND MANGANESE-54 IN PERIPHYTON

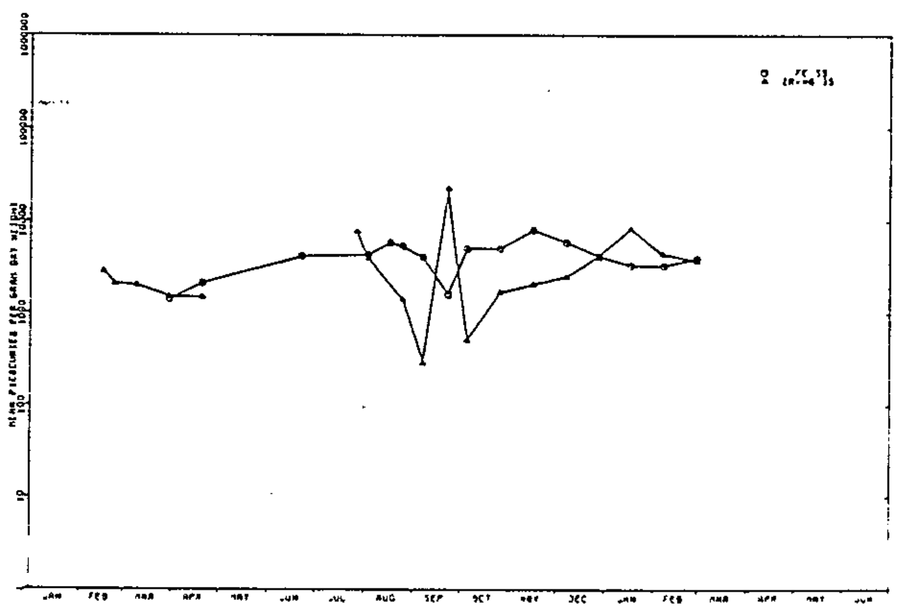

FIGURE 9. CONCENTRATIONS OF IRON-59 AND ZIRCONIUM-NIOBIUM-95 IN PERIPHYTON

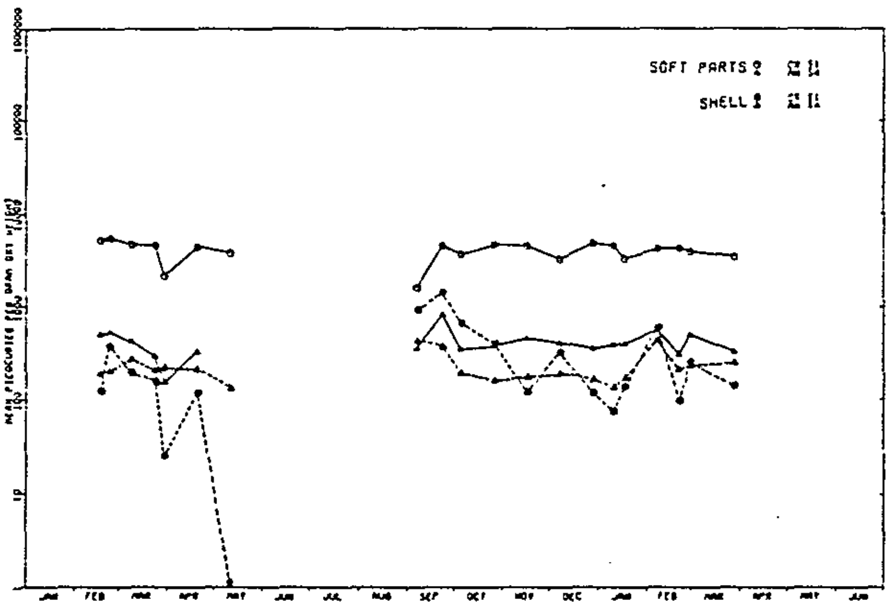

FIGURE 14. CONCENTRATIONS OF CHROMIUM-51 AND MANGANESE-54 IN LIMPET SOFT PARTS AND SHELL, Fisherola nuttalli

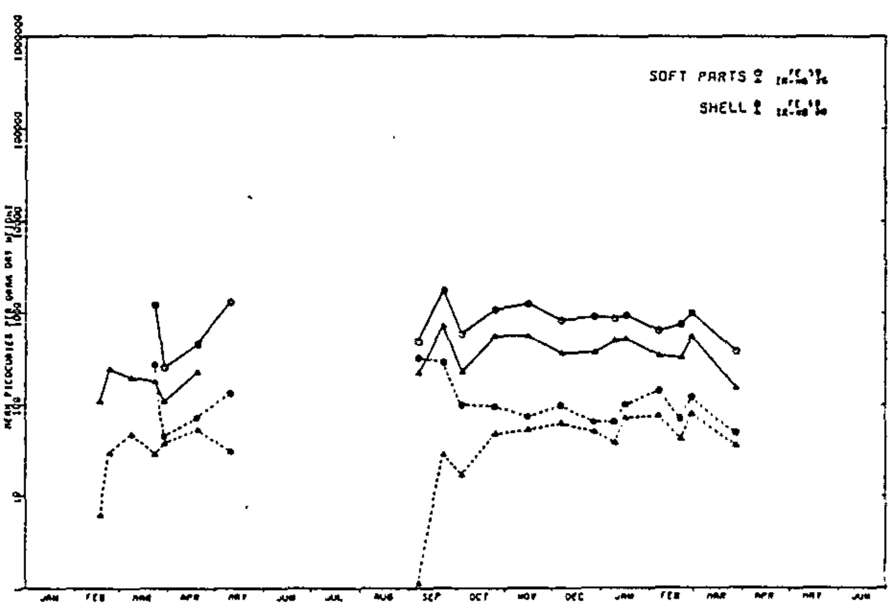

FIGURE 15. CONCENTRATIONS OF IRON-59 AND ZIRCONIUM-NIOBIUM-95 IN LIMPET SOFT PARTS AND SHELL, Fisherola nuttalli

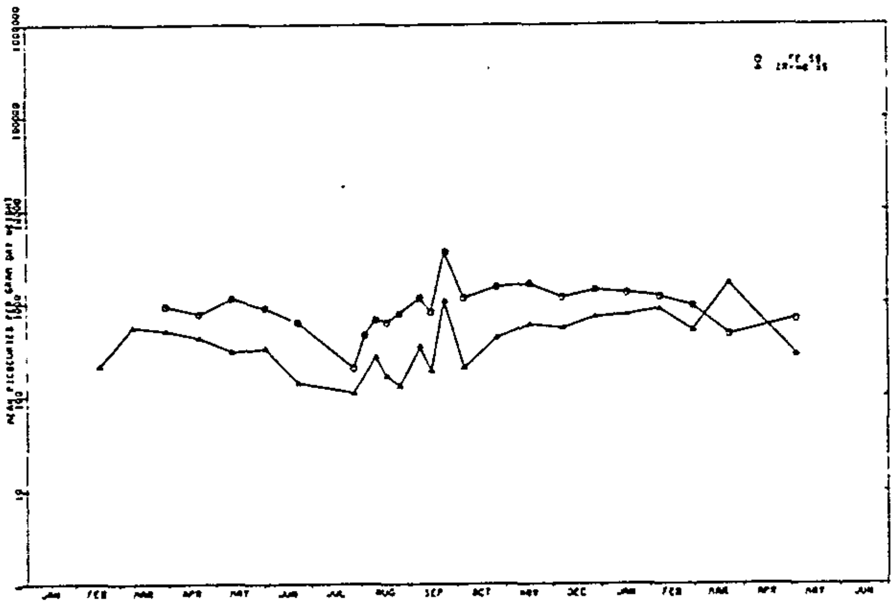

FIGURE 12. CONCENTRATIONS OF IRON-59 AND ZIRCONIUM-NIOBIUM-95 IN CADDISFLY LARVAE, HYDROPSYCHIDAE

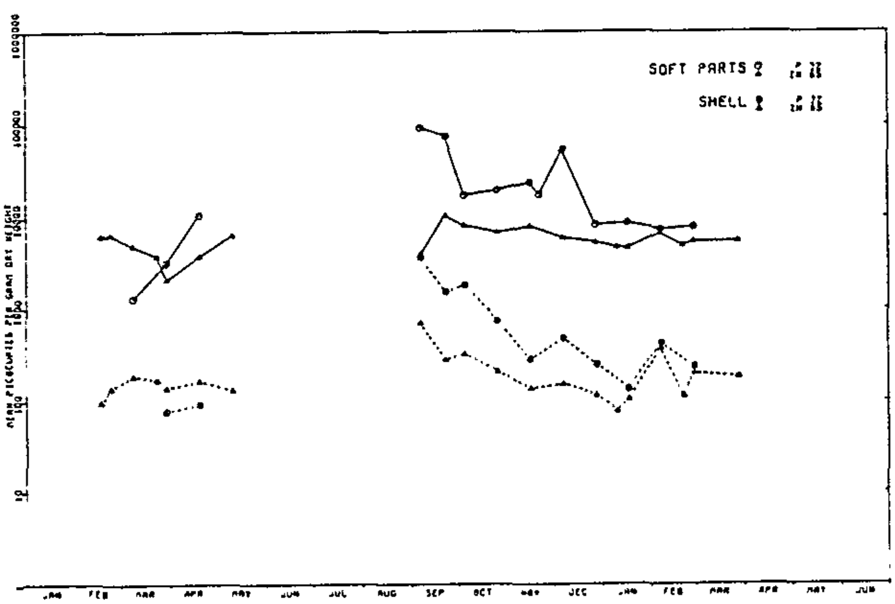

FIGURE 13. CONCENTRATIONS OF PHOSPHORUS-32 AND ZINC-65 IN LIMPET SOFT PARTS AND SHELL, *Fisherola nuttalli*

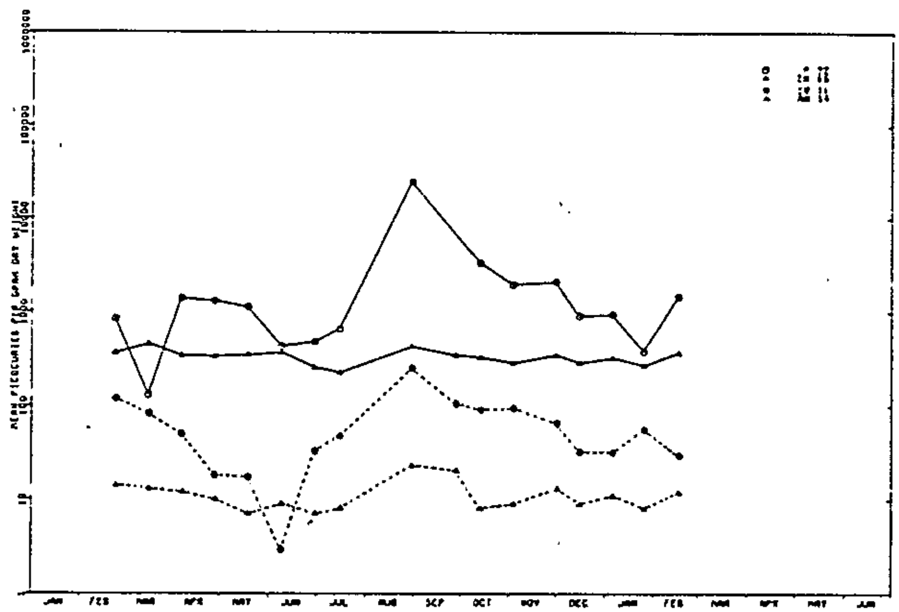

FIGURE 18. CONCENTRATIONS OF PHOSPHORUS-32, ZINC-65, CHROMIUM-51, AND MANGANESE-54 IN LARGESCALE SUCKER CARCASS, Catostomus macrocheilus

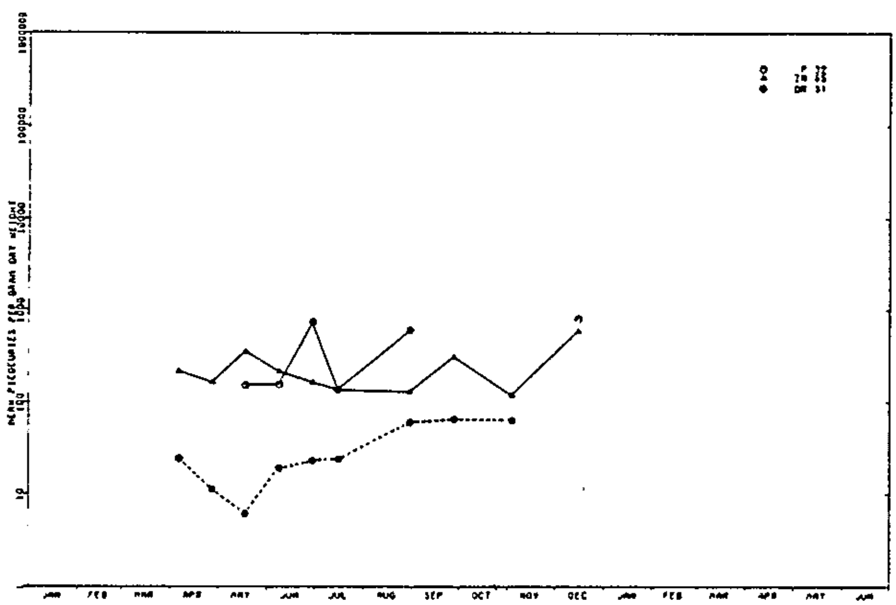

FIGURE 19. CONCENTRATIONS OF PHOSPHORUS-32, ZINC-65, AND CHROMIUM-51 IN NORTHERN SQUAWFISH CARCASS, Ptychocheilus oregonensis

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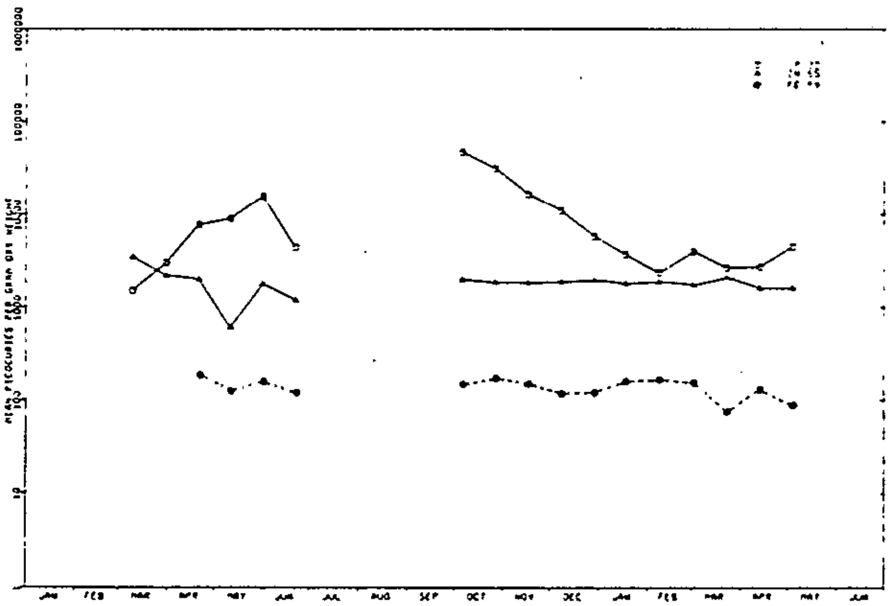

FIGURE 16. CONCENTRATIONS OF PHOSPHORUS-32, ZINC-65, AND IRON-59 IN JUVENILE REDSIDE SHINERS, *Richardsonius balteatus*

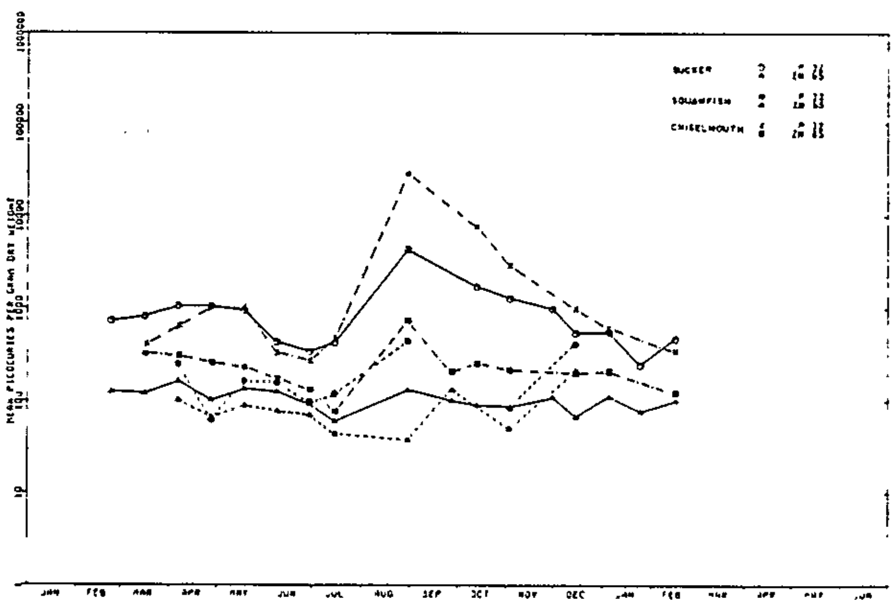

FIGURE 17. CONCENTRATIONS OF PHOSPHORUS-32 AND ZINC-65 IN MUSCLE OF LARGESCALE SUCKER, *Catostomus macrocheilus*, CHISELMOUTH, *Acrocheilus alutaceus*, AND NORTHERN SQUAWFISH, *Ptychocheilus oregonensis*

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temperature (Fig. 3) with the seasonal fluctuation in radionuclide burden in the organisms shows this relationship. In general, minimum levels for most radionuclides in both plant and animal forms occurred during the late spring and early summer, and were primarily the result of maximum dilution of reactor effluents by the annual freshet. High turbidity, which limits autotrophic metabolism through the restriction of light, and increased quantity of suspended sediments offering more sites for isotope adsorption also contribute to the lower concentrations of radionuclides during periods of high flow.

The season of maximum radionuclide burden was different in plant and animal forms. Highest levels in animals usually occurred in late summer and early fall when river flows were decreasing and water temperatures were at the yearly maximum. The slight increase in radionuclide concentrations accompanying the spring rise in river temperatures was reversed by dilution of the annual runoff. Concentrations in the autotrophic organisms, plankton and periphyton, showed maximums in late fall and winter when both flow and temperature were at a minimum, and radionuclide concentrations in the water at a maximum.

Phosphorus-32 was generally the most abundant nuclide in the biota and showed a relatively large seasonal variation. Differences between the annual minimums and maximums ranged from 10 to 100 fold, and greatest seasonal changes occurred in large fish tissues. Concentrations of 200,000 pCi phosphorus-32/g dry wt were observed in periphyton and there was a general decrease in concentration with progressively higher trophic levels. The comparatively high levels of phosphorus-32 in river organisms are due to the low concentrations of stable phosphorus in the river water

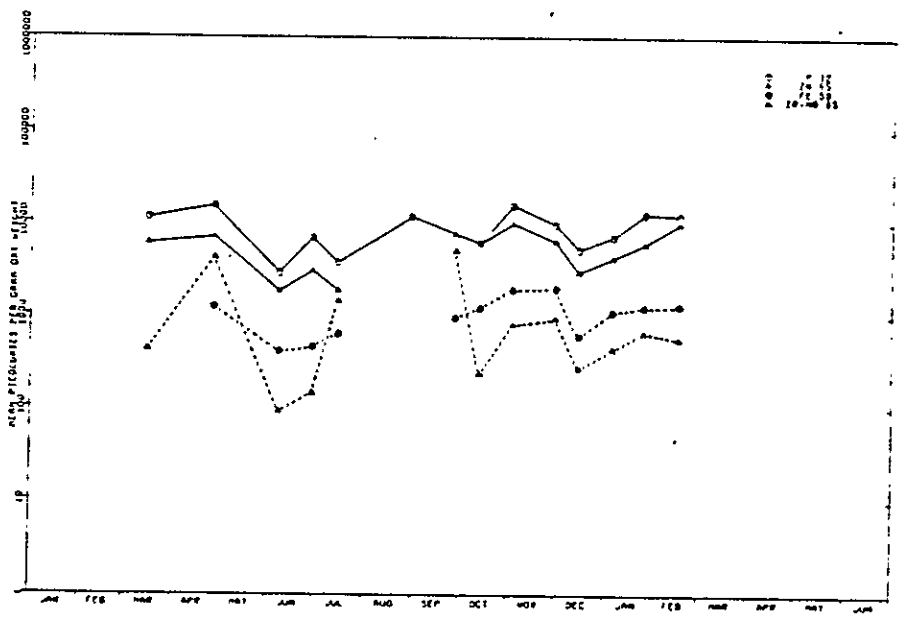

FIGURE 20. CONCENTRATIONS OF PHOSPHORUS-32, ZINC-65, IRON-59, AND ZIRCONIUM-NIOBIUM-95 IN LARGESCALE SUCKER GUT CONTENTS, Catostomus macrocheilus

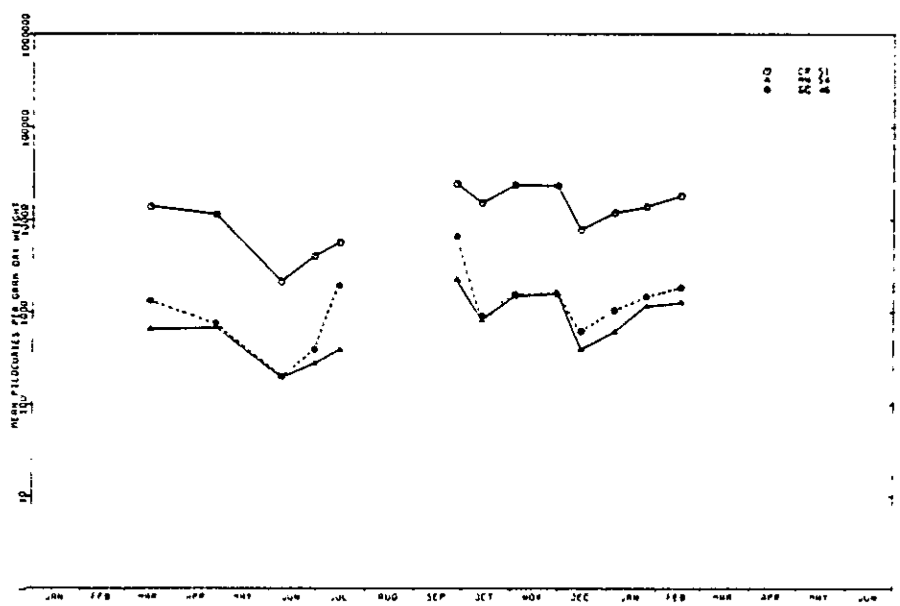

FIGURE 21. CONCENTRATIONS OF CHROMIUM-51, MANGANESE-54, AND SCANDIUM-46 IN LARGESCALE SUCKER GUT CONTENTS, Catostomus macrocheilus

in trophic level, with very little seasonal fluctuation evident in fish. This is partly due to the long effective half-life of this element in fish. In rainbow trout, a half-life of 134 days has been reported (19), and is probably similar in other fishes.

Chromium-51 was the most abundant radionuclide in plankton and periphyton, was nearly as plentiful as phosphorus-32 and zinc-65 in the invertebrates, but was absent or infrequently observed in fish muscle. It was consistently found in fish "carcass," the portion that included the outer surface of the animals. Radiochromium is of little biological importance as a metabolite, but because of its relative abundance in the water (Fig. 2), much is adsorbed on the outer surfaces of river organisms. Its seasonal variation is much less than that of phosphorus-32 with the maximum concentration usually less than 20 times the minimum. The gut contents of the invertebrates, caddisfly larvae and limpets, are composed largely of algae and detritus, and contribute significantly to the chromium-51 burden in these animals.

The seasonal pattern of concentrations of manganese-54, iron-59, and zirconium-niobium-95 were similar with the maximum usually less than 20 times the minimum. Both iron-59 and manganese-54 showed little seasonal change in fish and zirconium-niobium-95 was not found often enough to show seasonal trends.

Total Beta Comparisons

The usefulness of total beta activity measurements is of limited value because of the varying influence of contributing radionuclides.

(ca 0.01 ppm PO_4) and to its high biological concentration (18), thus enhancing the relative amounts of radioactive phosphorus.

Zinc-65 was generally the second most abundant radioelement in the biota. Its concentrations and seasonal variations were almost identical to those of phosphorus-32 in net plankton. Its concentrations in periphyton, composed mainly of sessile plant forms, was appreciably lower than that of phosphorus-32. The net plankton, primarily phytoplankton, is derived to a large extent from the river upstream from the reactor outfalls, although the sloughing of radioactive periphyton downstream from the reactors contributes to the radionuclide burden of the plankton that was collected in this study. The exposure of the plankton to the reactor effluents is relatively brief and equal to the few hours necessary for the river to travel from the points of effluent discharge to the collection site. Plankton exposure to radionuclides is confined to those contained in the water mass in which it is traveling and therefore reflects short term fluctuations in the radioactivity of the water. Much of its radioactivity is adsorbed to the outer surface and not assimilated by the algal cells. Although both the plankton and periphyton have large surface to volume ratios making surface adsorption an important route of uptake, the periphyton, because of its fixed position acts as an integrater and reflects a smoother seasonal pattern of accumulation. A definite summer decrease in zinc-65 in periphyton was observed in 1963-64 (10), but is not as evident in the few samples available during the high water period of the present study.

Seasonal variations in zinc-65 concentrations decrease with increase

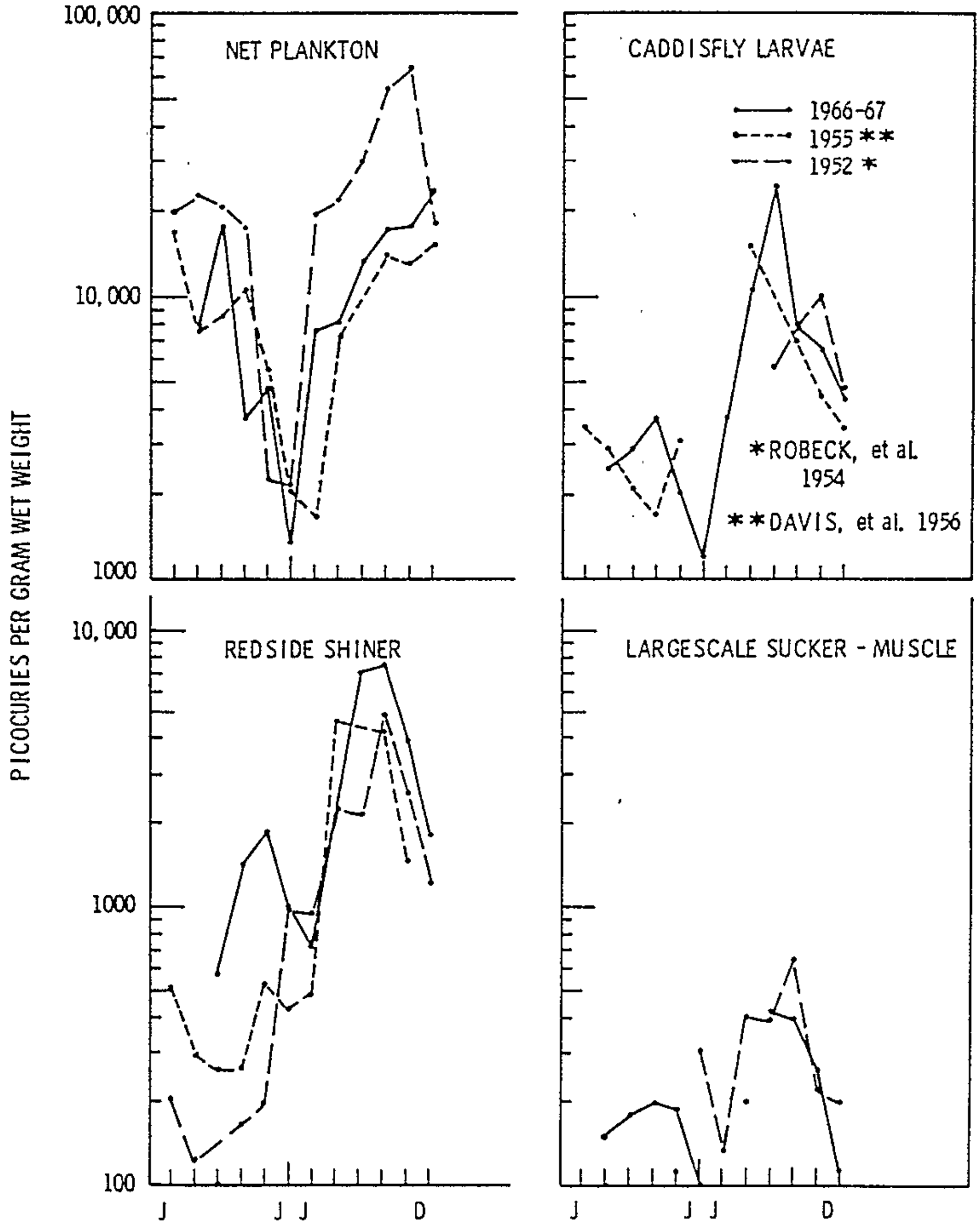

FIGURE 22. COMPARISON OF TOTAL BETA CONCENTRATION IN RIVER ORGANISMS FOR THE YEARS 1952, 1955, AND 1966-67

Total beta analyses do offer a basis, however, for comparing measurements obtained prior to 1956 with those of this study. The direct comparison of the amounts of beta radioactivity found in the present study with that of earlier investigations is hampered by (1) the number of operating reactors, and hence effluent output, changed with time, (2) modifications in cooling water treatment that altered the character of the effluent in terms of amounts and relative proportions of certain radionuclides, and (3) changes and refinement of beta measurement in the biological materials.

Figure 22 presents the seasonal concentrations of total beta activity in four organisms during 1952, 1955, and 1966-67.

In general, seasonal fluctuations and levels were similar for the plankton, caddisfly larvae, and largescale sucker muscle. This is especially remarkable when one considers the changes which occurred in the River during this period as mentioned in the Introduction.

Net plankton concentrations were generally higher in 1952 and the time of the lowest concentrations was in June, a month earlier than in 1955 and 1966-67. Data for caddisfly larvae and largescale sucker are not complete enough for comparison of seasonal trends. Similar patterns, however, and levels of activity are evident where comparison is possible.

Trends in the concentrations of total beta activity in the redside shiners, however, appeared to be different during the January to June period of the three years, but similar in late summer and fall. Seasonal low values occurred at successively later dates; in February in 1952, March and April in 1955, and mid-summer in 1966-67.

Figure 23 shows the total beta concentrations in several representative species at four seasons for the years 1955 and 1966-67. Differences between species is greatest during the first quarter of the year when water temperature and flow are near the minimum; least differences occur during the second and third quarters of the year when higher temperatures enhance the metabolism of many of the animal species, and river flows are declining and approaching the annual minimum . A decrease in activity with progression up the trophic levels is very apparent in the first half of the year. This relationship changes in the third quarter when the activities in many of the animals approach or exceed the concentrations in net plankton.

In spite of the changes in reactor operation and sample analyses that limit the direct comparison of quantities of beta activity found in this study with that of earlier investigations, one would expect a greater change with time than is indicated in Fig. 23. With most organisms and at all seasons of the year, the concentrations of beta activity in 1966-67 are much the same as that of 1955. In 1955, eight reactors were operating, and only five in 1966-67. This would seem to indicate that the accumulation of radioactive materials by the river biota is not directly related to the number of operating reactors, but to the total radioactivity in the effluents.

Gamma Comparisons

Analytical capabilities for measuring concentrations of gamma radionuclides have been perfected since results of many of the early River studies were completed. Hence, relatively few gamma measurements

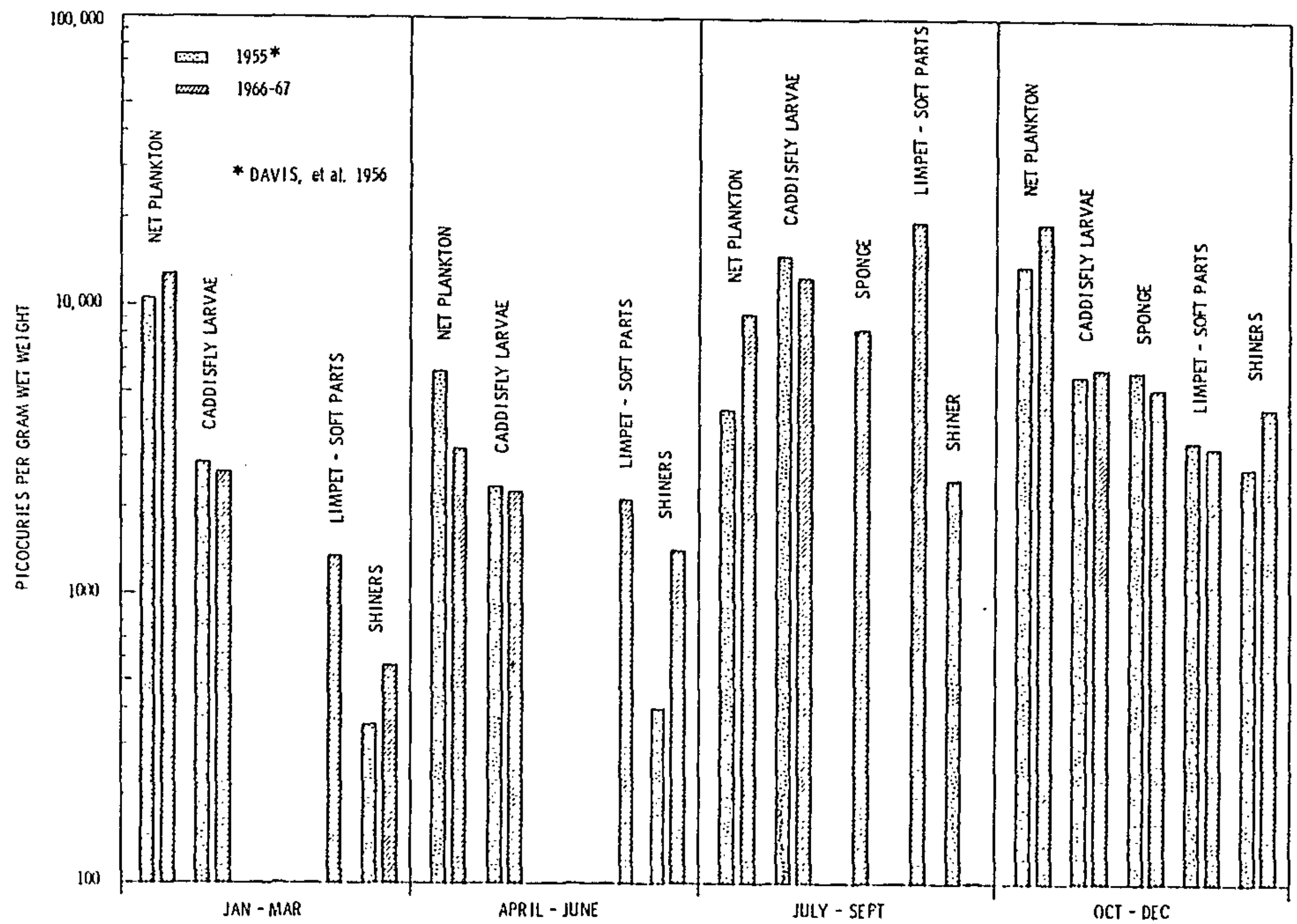

FIGURE 23. INTERSPECIES COMPARISON OF BETA ACTIVITY CONCENTRATION IN COLUMBIA RIVER ORGANISMS, 1955* AND 1966-67

TABLE 1. Comparison of Concentrations of Gamma Emitting Radionuclides in Columbia River Organisms 1957-67

		pCi/g wet weight						
		Plankton	Sessile Green Algae	Sponge	Caddisfly Larvae	Limpet Soft Parts	Limpet Shell	Minnows
^{46}Sc	1957[1]	--	1730	94.7	70.6	--	--	0.706
	1967[2]	5690	3020	2130	968	87	475	0 [3]
^{51}Cr	1957	--	7900	4580	6000	--	--	372
	1964[4]	59500	43400	10200	3590	1940	1080	--
	1965[4]	28400	32900	16000	4890	2260	1350	--
	1967	12600	10160	5060	3030	696	1060	17.6[3]
^{54}Mn	1957	--	1030	--	79.1			
	1967	791	1080	603	447	136	359	0 [3]
^{59}Fe	1957	--	1640	--	--	--	--	--
	1967	1250	1360	860	537	260	274	28.4[3]
^{60}Co	1957	--	155	11.6	1.72	--	--	--
	1967	41	456	0	7	80	31	0 [3]
^{65}Zn	1957	--	12300	1460	1980	--	--	762
	1964	14000	8870	3070	1970	2820	658	--
	1965	1910	3250	2500	1770	1360	346	--
	1967	4580	2050	1910	1790	1560	435	237[3]
^{95}Zr-Nb	1957	--	1790	--	66.3	--	--	--
	1967	953	380	553	156	109	13	0
^{140}Ba	1957	--	901	--	42.2	--	--	--
	1967	1910	459	510	367	96	117	0 [3]

are available prior to 1960. Analysis of these early data are also subject to the same qualifications mentioned in the previous section comparing total beta activity.

Comparison of the concentrations of gamma emitting radionuclides are given in Table 1. All data presented in this table were collected during approximately the same time of year (August-October) and from the same general area of the river immediately downstream from 100-F area. Similar to the total beta measurements, no consistent time related trends in the concentrations of specific radionuclides is apparent.

Effects of Reactor Shutdown

The shutdown of the reactors in July and August of 1966 produced marked results in the concentrations of radionuclides in the biota. Concentrations of radionuclides decreased rapidly to varying levels. Chromium-51 and phosphorus-32 decreased two to three orders of magnitude in the lower trophic levels; phosphorus-32 levels were not measurable within about seven days after shutdown and chromium-51 levels were below detection limits after about five weeks. The declines in concentrations of zinc-65, manganese-54, and iron-59 were much less and measurable amounts remained present at all times. Decreases of an order of magnitude, however, were common in most organisms. In fish, phosphorus-32 was lost rapidly and zinc-65 more slowly.

The fact that some elements remained present in measurable quantities suggests that quantities of these isotopes were still being contributed to the river. Two sources contributed radionuclides to the water during shutdown: (1) a small amount of certain nuclides from water pumped through the reactor cooling systems, and (2) release of radionuclides sorbed to the river sediments. Estimates of the percentage decrease of several radionuclides from McNary Reservoir sediments due to leaching and scouring during the shutdown period have been measured (20). These range from 20% for chromium-51 and scandium-46 to 50% for zinc-65 and 75% for manganese-54 and cobalt-60. Cycling of these elements probably provided the radionuclides which were present in the biota during shutdown. All organisms rapidly accumulated the radionuclides following resumption of reactor operation and near equilibrium concentrations in most organisms were approached within two or three weeks. Figures 24 and 25 present typical curves showing the influence of the reactor shutdown on radionuclide concentrations in net plankton and shiners. A fuller description has been published (11).

The changes in zinc-65 concentration, due to reactor shutdown, were measured in estuarine juvenile starry flounders (<u>Platichthys stellatus</u>) near the mouth of the Columbia River (21). After correction of about 19 days for river flow time between Hanford and the river mouth, a lapse of nearly a month occurred before a significant decline in zinc-65 concentration was noted in the starry founder. This is comparable to the time lapse before a marked reduction in zinc-65 was evident in shiners collected near the reactors (Fig. 25). After the reactors resumed operation, fish at both sites approached a "normal" zinc-65 concentration within two weeks.

TABLE 1. (continued)

		pCi/g wet weight						
		Plankton	Sessile Green Algae	Sponge	Caddisfly Larvae	Limpet Soft Parts	Limpet Shell	Minnows
^{140}La	1957	--	3270	1230	347	--	--	--
	1964	5900	1610	950	223	73	113	--
	1965	2010	1760	1330	322	107	107	--(3)
	1967	4630	2400	2400	656	333	379	0
^{239}Np	1957	--	2690	401	311	--	--	--(3)
	1967	3010	1750	1080	384	79	173	0
^{32}P	1957	--	66000	4460	24300	--	--	24000
	1966			3270	6560	3790	988	7110
	1967		12800	15100	28200	19000	2310	

(1) Davis, J. J., et al. 1958, Ref. 7.

(2) Present study September 1967 data.

(3) Present study September 1966 data

(4) Cushing, C. E. and D. G. Watson, 1966, Ref. 9.

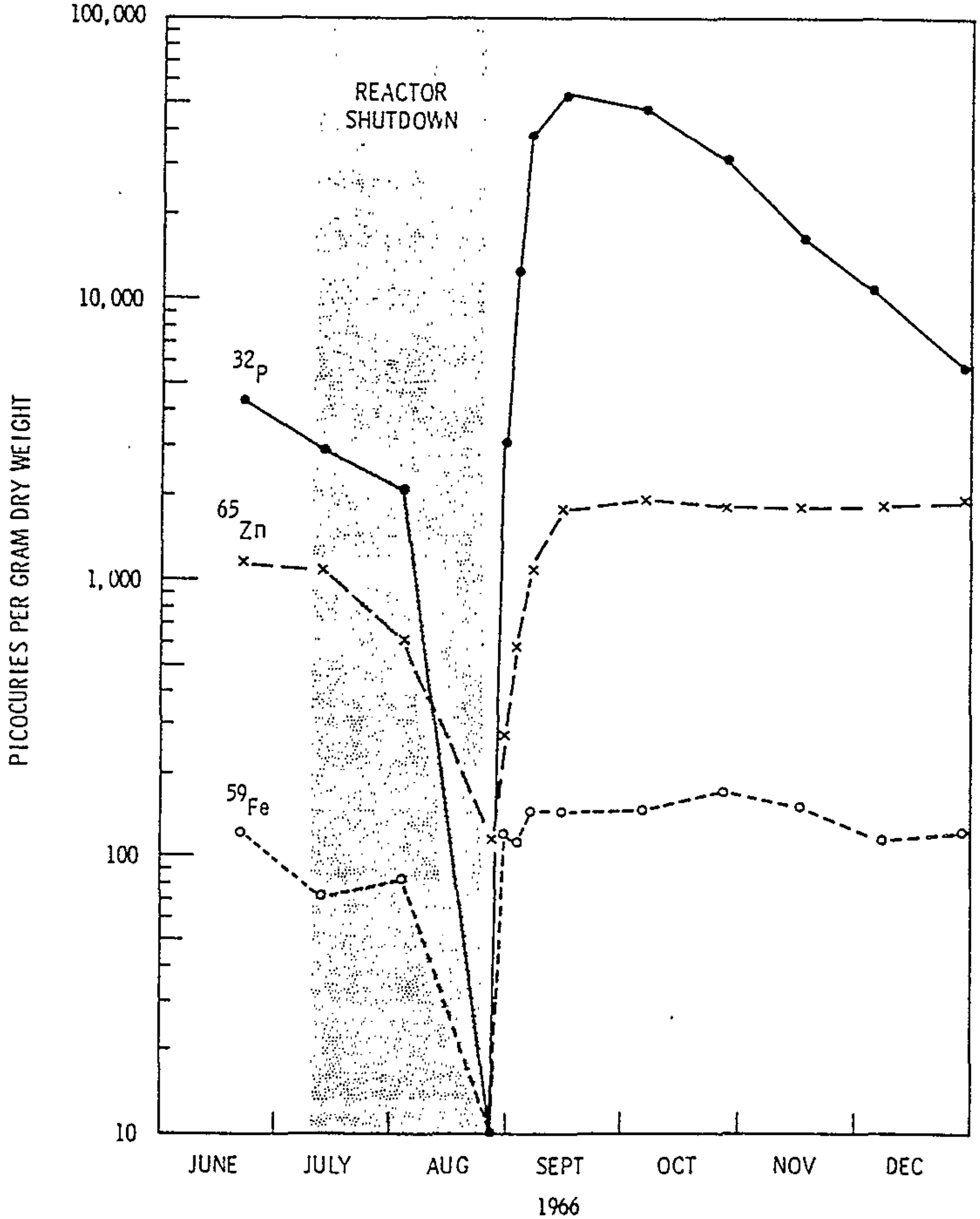

FIGURE 25. RADIONUCLIDE CONCENTRATION IN SHINERS (*Richardsonius balteatus*) DURING AND AFTER REACTOR SHUTDOWN

9 2 1 2 3 7 7 2 1 2 3

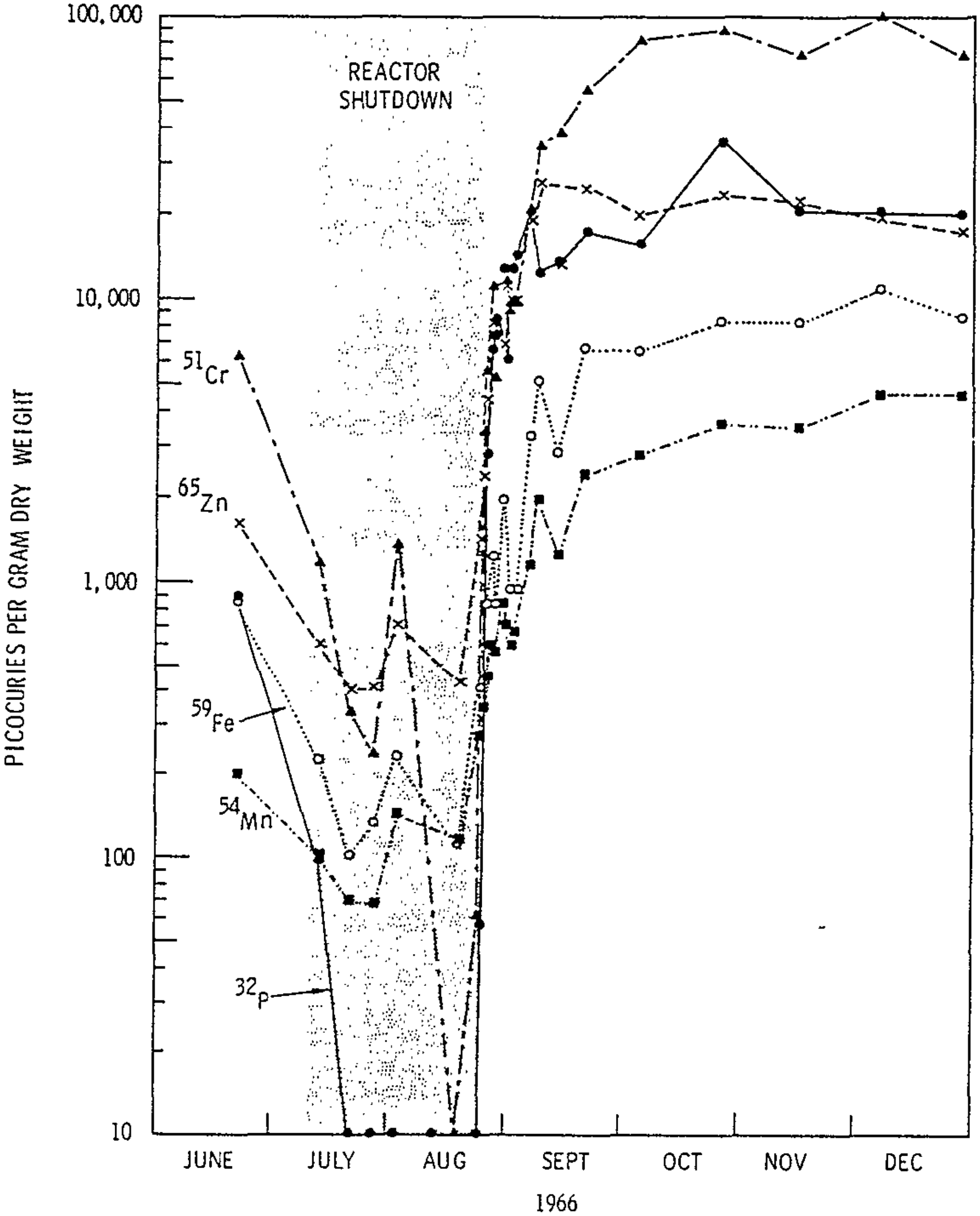

FIGURE 24. RADIONUCLIDE CONCENTRATION IN NET PLANKTON DURING AND AFTER REACTOR SHUTDOWN

9 2 1 2 1 7 7 2 1 2 2

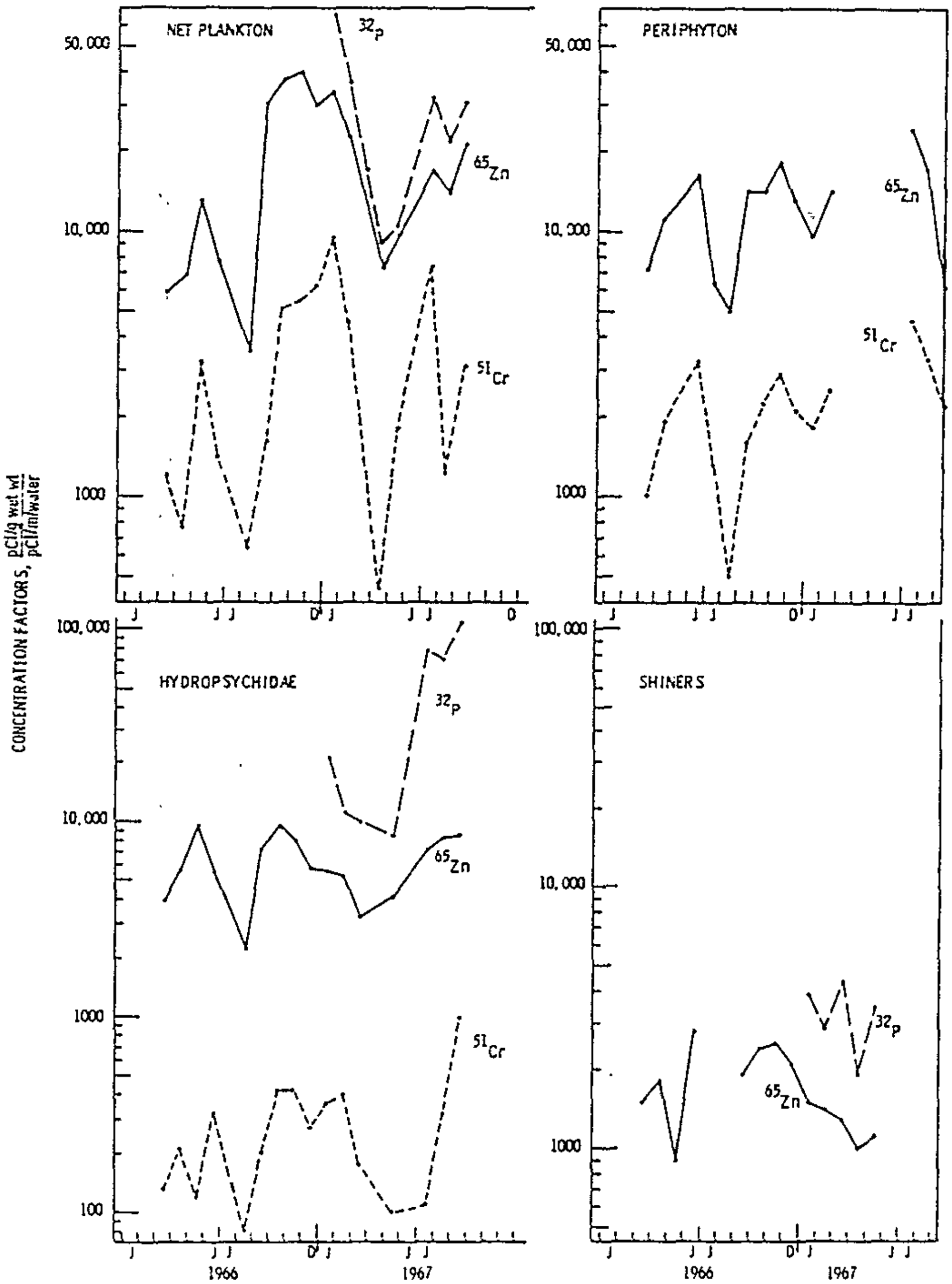

FIGURE 26. CONCENTRATION FACTORS FOR COLUMBIA RIVER ORGANISMS, 1966-67 (NOTE SCALE DIFFERENCE)

Concentration Factors

The concentration factor (CF) is an expression used to estimate the degree to which an organism concentrates a particular radionuclide or element from the ambient water. It is calculated by dividing the pCi/g wet weight of the organism by the pCi/ml of the same radionuclide in the water. There has been a good deal of discussion as to the validity of this measurement and a discussion of its present status has been published (22).

Figure 26 presents the seasonal variations in the concentration factors of zinc-65, chromium-51, and phosphorus-32 in net plankton, periphyton, caddisfly larvae, and redside shiner.

Analysis by Species

Zinc-65 and phosphorus-32 were concentrated to a higher degree by net plankton than chromium-51. Zinc-65 CFs ranged from 3500 to 40,000 with low values occurring in summer and high values in winter. Phosphorus-32 values ranged from 9000 to 68,000, and chromium-51 from 440 to 9400. Zinc-65 and phosphorus-32 CFs ranging from 300 to 19,000 and from 5000 to 118,000, respectively, have been reported for net plankton in the Columbia River in 1963-64 (23). These values include those of the present study for phosphorus-32, but are lower for zinc-65.

The seasonal variations observed in the plankton were also apparent in the other organisms sampled. It is probably related more to the seasonal dilution of radionuclides in the water by high summer flows than to seasonal changes in metabolic activity. The latter would be expected

Analysis by Radionuclide

Zinc-65 CFs were highest in the autotrophic plankton and periphyton and decreased through the caddisfly larvae and shiners.

Phosphorus-32 CFs were higher in caddisfly larvae, though only slightly lower in net plankton; lowest values were found in the shiners. No values are available for periphyton since it was unavailable during the period for which we have phosphorus-32 water data.

Concentration factors for chromium-51, a biologically unimportant element, were of a similar order of magnitude for net plankton, periphyton, and caddisfly larvae; levels were not measurable in shiners. Lower values would be expected for this adsorbed element by caddisfly larvae because of their smaller surface to volume ratio as compared to those of the algae.

These data conflict somewhat with the usually observed increase in CFs as one moves up the trophic level, i.e., from autotrophs to herbivores.

Comparison with Total Beta CFs (1955)

Comparison of CFs for total beta, the only data available from the earlier report (4), with those of phosphorus-32 from the present study are tenuous because of the unknown contribution of other beta emitters to the earlier data. Nevertheless, the majority of the activity was probably due to phosphorus-32, so comparative data are given in Table 2.

to produce higher CFs during warm weather in summer and low CFs in colder months. This pertains to that radioactivity which is incorporated by metabolic processes governed by temperature and not to that radioactivity which is adsorbed and is a function of surface to volume ratios. Chromium-51, for instance, is not biologically important and is accumulated by adsorption. Zinc-65 is accumulated mainly by adsorption by plankton and periphyton but by food-chain absorption by caddisflies.

Zinc-65 CFs in periphyton were again higher than those of chromium-51. Values for zinc-65 ranged from 5000 to 24,000; for chromium-51 from 500 to 4300, approximately an order of magnitude difference.

Phosphorus-32 CFs in Hydropsychidae were about an order of magnitude higher than those for zinc-65, which, in turn, were an order of magnitude higher than CFs for chromium-51. Values ranged from 8400 to 100,000 for phosphorus-32, 22,000 to 97,000 for zinc-65, and from 800 to 9900 for chromium-51. Concentration factors of 100,000 for phosphorus, 2,000 for chromium, and 40,000 for zinc have been reported for fresh water invertebrates (24). These are within agreement with the present results.

Concentration factors for both zinc-65 and phosphorus-32 were considerably lower in shiners than in the net plankton, periphyton, or caddisfly larvae. Concentration factors for zinc-65 ranged from 900 to 2800; for phosphorus-32 from 1900 to 4200. Concentration factors of 100,000 for phosphorus, 200 for chromium, and 1000 for zinc have been reported for freshwater fish (24). Zinc-65 CFs for the shiners agree with these data, but phosphorus-32 CFs are much different. Chromium-51 levels were not measurable in shiners.

particularly Na and K, have provided questionable results, so that the values in Table 3 should be regarded as approximations.

TABLE 3. Specific Activities, µCi/g

	P	Na	K	Zn	Fe	Mn	Cr	Co
Net plankton	6.03	2.24		24.6	0.624	3.28	1830	
Periphyton	4.93			20.3	0.764	4.85	1570	
Sponge	1.17			14.5	0.417	2.44	972	
Caddis larvae	2.76	0.185		13.3	0.701	3.35	459	
Limpet								
shell	1.47	0.0969		8.50	1.59	2.53	35.7	
soft parts	0.757	0.840		18.5	0.107	3.28	137	5.90
Shiner	0.810	0.162		16.8	4.56		> 48.3	
C.S. sucker								
muscle	0.134	0.130	0.00158	5.75		0.827	> 0.340	
carcass	0.107	0.176	0.00307	6.71		0.624	60	0.375
Gut cont.	2.96			11.1	0.0872	1.83	520	2.10

Table 4 lists some specific activities reported in the literature for comparison with those in Table 3.

TABLE 4. Specific Activities

	Zn	P	Mn	Reference
Clam, soft parts	250		25	26
Clam, soft parts			3000	27
Fish, whole		0.5		8
Chironomid larvae		4		8

TABLE 2. Concentration Factors - Total Beta (1955), Phosphorus-32 (1966-67)

	Total Beta		Phosphorus-32
	Average	Maximum	Range
Net plankton	1600	2600	9000 - 68,000
Caddisfly	1300	3000	8400 - 108,000
Shiners	470	900	1900 - 4200

In all cases, data from this study showed marked increases over results from the earlier studies. This may be partially related to improved radioanalytical techniques.

Specific Activity

The specific activity of an element in an organism (ratio of radioactive to stable atoms) is perhaps the best measurement of the ecological demand for that element, assuming that certain limitations are met (25). Use of specific activities allows one to predict expected contamination levels in organisms if the stable concentrations are known for the organism and the media and also the expected environmental levels of contamination.

Table 3 presents the specific activities for eight elements in representative Columbia River biota sampled in November and December 1966. Since the stable element analyses for these data were done, we have become aware that the techniques used for analyzing certain elements, view of the environmental and technological changes which have occurred, this was quite unexpected.

4. No consistent time related trends were apparent in the gamma activity measurements between the present data and that collected prior to 1956.

5. The shutdown of all reactors for several weeks during this study produced marked changes in the radionuclide concentrations in the biota. Levels of most nuclides dropped several orders of magnitude and many were undetectable. Measurable amounts of certain isotopes were present from radioactivity recycled from the sediments or small amounts coming from the reactor effluents. After the reactors resumed operation, levels increased rapidly to pre-shutdown levels or higher.

6. Concentration factors were highest for the biologically important radionuclides phosphorus-32 and zinc-65 and were highest in the primary producers, net plankton and periphyton, and decreased in higher trophic levels.

Selected specific activities are presented and compared with values from the literature.

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The closest agreement is between the specific activities for phosphorus in the chironomid larvae (4) and that for the caddis larvae (2.76). For comparison, the specific activity of Columbia River water in January 1964, was 3.8 for phosphorus and 225 for zinc.

CONCLUSIONS

A study of radionuclide concentration levels in Columbia River biota was undertaken to (1) define the interspecies and seasonal variations in the concentration of several of the more biologically important radionuclides, and (2) update the findings of some of the earlier investigations.

1. Concentrations of radionuclides in the Columbia River water are generally inversely proportional to flow levels, with highest values in winter and lowest in summer. This is related to dilution by seasonal runoff.
2. Concentrations of most radionuclides in the biota followed a pattern of high levels in winter followed by low concentrations in summer. There were often indications of an increase in concentration just after levels started to decline in spring. This may be attributed to improved light and temperature regimes which favored metabolic uptake; this increase, however, was soon overwhelmed by the dilution of radionuclides by the spring runoff.
3. A remarkable similarity in total beta concentration was observed between the present data and that collected prior to 1956. In

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